Techno-economic analysis of geopolymer production from the coal fly ash with high iron oxide and calcium oxide contents

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Abstract: In this work, we firstly examined the technical feasibility of geopolymer synthesis from the coal fly ash with high iron oxide (48.84 wt.%) and calcium oxide (22.15 wt.%) contents. The heat resistance of geopolymer was represented by the dry weight loss which ranged from 2.5 to 4.9% and was better than that (11.7%) of OPC. However, the high iron oxide content made the acid resistance (13 - 14%) of geopolymer inferior to OPC. The economics of geopolymer production changes significantly upon the variation in the arrangement of material use and geopolymer price. The costs of Na_2SiO_3 and NaOH and the benefit of geopolymer selling were the major factors affecting the economic feasibility of geopolymer production. When the Na_2SiO_3 price was around 400 USD/ton, the geopolymer production will be profitable even if the geopolymer price was as low as 50 USD/ton. It is possible to improve the economics of geopolymer production by varying the arrangement of material use while not impairing the performance of geopolymer.

Keywords: Geopolymer; Coal fly ash; Ordinary Portland cement; Circular economy; Cost-benefit analysis.

INTRODUCTION

Geopolymer is receiving an increasing attention from both academia and industry because it could be used to develop reduced-CO_2 construction materials and serve as an alternative or replacement to ordinary Portland cement (OPC) (Provis and Van Deventer, 2009). The pressure of energy cost and climate change has motivated extensive development and application of geopolymers. Commercial geopolymers have been applied to railway sleepers, sewer pipes, building products, fireproof wall panels, protective coatings, masonry materials, *etc*. (Glasby et al., 2015; Wallah and Rangan, 2006).

Geopolymer is produced through geopolymerisation by mixing aluminosilicate with alkaline solution which consists of alkali metal silicate solution and alkali metal hydroxide. A combination of sodium

hydroxide (NaOH) and sodium silicate (Na_2SiO_3) is the most common alkaline solution used in geopolymerisation. The geopolymerisation process typically involves three steps, *i.e.* aluminosilicate dissolution, gelation and structure reorganization, and polymerisation and hardening (Santa et al., 2013; Xu and Van Deventer, 2000). Geopolymers mainly consist of amorphous or semi-crystalline polymeric structures of silicon-oxygen-aluminium bonds that are formed by alternating silicon and aluminium tetrahedra joined together by sharing all the oxygen atoms (Heah et al., 2012). The properties and performance of geopolymer can be affected by a variety of factors including SiO_2/Na_2O mole ratio, temperature, curing time, types of alkaline solution, *etc.* (Blissett and Rowson, 2012; Part et al., 2015; Sukmak et al., 2013; Xu and Van Deventer, 2000). Additionally, geopolymerisation is affected by the type of source materials that vary in structures and contents (Hardjito et al., 2004; Temuujin et al., 2009; Yip et al., 2008). This means that it is always desirable to examine the geopolymerisation process for a specific type of source material which bears a unique structural or compositional feature.

Coal fly ash is the main raw material for geopolymer production because of its widespread availability and low cost (Ferone et al., 2011). Coal fly ash-based geopolymer production also provides a sustainable way for the disposal of coal fly ash that remains a worldwide challenge (Blissett and Rowson, 2012; Ferone et al., 2011). The chemical compositions of coal fly ash are highly contingent upon its origin (Reijnders, 2005). Although an existing study has examined the compressive strength of geopolymer derived from the coal fly ash with a medium iron oxide content (13.2 wt.%) (Rickard et al., 2011), systematic information about the influences of the various factors on the heat resistance and acid resistance of geopolymer derived from the coal fly ash with high iron oxide and calcium oxide contents is still lacking.

In this work, we firstly test the technical feasibility of synthesizing geopolymer using coal fly ash with

high iron oxide (48.84 wt.%) and calcium oxide (22.15 wt.%) contents. A cost-benefit analysis (CBA) is conducted to identify the economic feasibility boundaries of geopolymer production by incorporating geopolymer production conditions. There have been limited studies (Abdollahnejad et al., 2015; Chan et al., 2015) assessing the economic feasibility of geopolymer production. The existing studies generally did not consider the time value of money and uncertainty in the analysis and the economic feasibility boundaries of geopolymer production are still unknown.

2. METHODOLOGY

2.1 Technical analysis

2.1.1 Materials

The coal fly ash was from a coal-based power generation plant in Singapore and has a brown colour appearance. For comparison, OPC was obtained from NUS Department of Civil and Environmental Engineering laboratory. Prior to wet treatment, the coal fly ash and OPC were dried in an oven at 105 °C for 24 hours. Both the coal fly ash and OPC were then sieved (Retsch AS200 digit) and it was found that around 60 wt.% of coal fly ash particles and more than 90 wt.% of OPC particles were smaller than 50 μm. The particles smaller than 50 μm were used for further analysis to reduce the incomparability of particle sizes between the coal fly ash and OPC. The sodium silicate (Na_2SiO_3) solution (Sigma-Aldrich Co.) contains 10.6% of Na_2O and 26.5% of SiO_2 and has a density of 1.39 g/ml (25 °C). Sodium hydroxide (NaOH) pellets (Sigma-Aldrich Co.) have a purity of ≥97.0%. Sulfuric acid (H_2SO_4) (Sigma-Aldrich Co.) is used for acid resistance tests and has a concentration of 99.999 wt% and a density of 1.840 g/ml (25 °C). The sulfuric acid is diluted to a 5 wt% sulfuric acid solution for experimental use.

2.1.2 Geopolymer synthesis

(a) Synthesis procedure

The NaOH pellets were first dissolved in the Na_2SiO_3 solution followed by the addition of DI-H_2O. The solution produced was then mixed with the coal fly ash by stirring. The mixture was poured into plastic tube moulds to produce geopolymer cylinders with a diameter of 1.5 cm and height of 3 cm. The tube moulds were covered with plastic films with small holes. The moulds were then put into an oven at 60 °C for 24 h for hardening. The geopolymers were used in the subsequent tests of heat resistance and acid resistance.

(b) Parameter investigation

The impacts of the amount of Na_2SiO_3 solution, mole ratio of SiO_2 to Na_2O in the alkaline solution, mass ratio of the coal fly ash to the alkaline solution, temperature and curing time on the heat resistance and acid resistance of geopolymer were studied. To study the effect of the amount of Na_2SiO_3 solution, the oxide compositions of coal fly ash from Andini et al. (2008) was used as a reference to determine the relative amount of Na_2SiO_3 solution, NaOH, and DI-H_2O based on 50 g coal fly ash. A mole ratio SiO_2/Al_2O_3 of 4 and mass ratio of fly ash to the alkaline solution of 3 were adopted in the reference case. Three other cases have 50%, 150% and 200% of the amount of Na_2SiO_3 solution in the reference case; the amount of DI-H_2O was adjusted correspondingly to keep the mass ratio of coal fly ash to the sum of alkaline solution and DI-H_2O to be 1.

To study the impacts of other parameters, the oxide compositions of the coal fly ash obtained from the XRF analysis of this work were used, and the mass ratio of coal fly ash (50 g) to the sum of alkaline solution and DI-H_2O was also 1. Experimental arrangements include: (1) four mole ratios of SiO_2 to Na_2O (1, 1.15, 1.3 and 1.45) in the alkaline solution was tested while the other parameters were kept constant (ash/alkaline solution mass ratio = 3, temperature = 60 °C, curing time = 24 h) to study the impact of the mole ratio; (2) four ash/alkaline solution mass ratios (2, 3, 4 and 5) were tested at the temperature of 60 °C and curing time of 24 hours to study the impact of the mass ratio; (3) four curing

times were tested at the temperature of 35 °C (time = 2.5, 8, 8.5 and 9 days), 60 °C (time = 12, 16, 20 and 24 h) and 80 °C (time = 12, 16, 20 and 24 h), respectively to study the combined effect of curing time and temperature. The curing times were decided by referring to some existing studies (*e.g.*, Santa et al. (2013) and Van Jaarsveld et al. (2002b)). However, for the curing time of 35 ^{o}C, we found that 2 days are not enough for the full drying of geopolymer, and then raised the during times to 8, 8.5 and 9 days, respectively. Details of the experimental arrangement are shown in Table 1. For each experiment batch, 3 samples were used for acid resistance test while 1 sample was used for heat resistance test.

2.1.3 Samples characterization

The metal oxide compositions in the coal fly ash were determined with the XRF analysis. The morphology of all the samples (*i.e.* coal fly ash, geopolymer and OPC) were investigated by scanning electron microscopy (SEM, JEOL 2872). The structural properties of samples were characterized by using a SHIMADZU XRD-6000 diffractometer. The geopolymer samples were also investigated by the KBr disk method using Fourier transform infrared spectroscopy (Bio-Rad FTS 3500).

2.1.4 Performance test

To characterize the heat resistance, the dry weight loss of geopolymer was measured based on the TGA analysis following ASTM E2550-11. To characterize the acid resistance of geopolymer, the weight loss of geopolymer after acid immersion was measured. The geopolymers were immersed in 5 wt.% sulfuric acid for 3 days and then DI-H_2O for 12 h to remove the acid remained in the geopolymers followed by 24-hr oven drying. Sulfuric acid has been commonly used to simulate the acid corrosion in sewer pipe systems where sulfuric acid could be generated bacterially from hydrogen sulfide (Wallah and Rangan, 2006). For comparison, hardened OPC (35 °C for 4 days) cylinders were tested for its heat resistance and acid resistance following the same procedures and methods as those

of geopolymers.

2.2 Economic Analysis

A Monte Carlo simulation-based CBA was conducted to evaluate the economic feasibility of coal fly ash-derived geopolymer in Singapore. The analysis is based on an assumed scenario where a geopolymer plant is built near (2 km) to the power plant and run for 20 years in Singapore. Around 55 tons of coal fly ash is produced every day in Singapore. Considering the ash/alkaline solution mass ratio of 3, the mass of geopolymer produced each day is around 70 ton. This implies the volume of geopolymer produced each day is around 30 m^3 under a density of 2.4 ton/m^3 (Steins et al., 2014). The cost components include the capital cost (land, plant construction, and equipment), operating & maintenance (O&M) cost, material (water, Na_2SiO_3, and NaOH) cost, and transport cost. The nominal land area is set to be 200 m^2 to include the fly ash and geopolymer storage area, production area, and office area. This is a conservative value in view of the daily volume productivity of 30 m^3. The cost of equipment is estimated based on online information (Alibaba, 2018). The powers of grinder, oven, and stirrer are set to be 20, 10, and 20 kW, respectively. The grinder and stirrer are operated 9 h each day while the operating time of oven depends on the curing time (Table 1). In general, one set of equipment is deployed. However, if the curing time is longer than 24 h, the number of the sets of ovens is equal to the days of curing to avoid over-accumulation of coal fly ash. It is assumed that there is around 10% auxiliary electricity consumption (AEC) by other activities (*e.g.*, office lighting, air conditioning, etc) (You et al., 2017). Considering relevant data regarding the O&M cost of geopolymer production is still lacking, the one related to cement production is used. This is a conservative assumption because a simpler procedure is involved in the production of geopolymer compared to that of cement which implies a lower O&M cost. The benefit components include geopolymer selling income and refuse disposal fee. The selling price of geopolymer is set to be like that of OPC. For each cost or benefit component, a nominal value is identified and serves as the mode

of a triangular distribution used in the Monte Carlo simulation. The lower and upper limits are assumed to be 70% and 130% of the nominal value, respectively. This Monte Carlo simulation-based method is used to account for the uncertainty underlying the economic analysis (You et al., 2016). A list of cost and benefit components is given in Table 2.

Net-present value (NPV) is used as the economic indicator

$$NPV = \sum_{t}^{LT} \frac{C_{it}}{(1+r)^t} - C_0 \quad (1)$$

where C_{it} is the net cash inflow in a year t; C_0 is the total initial investment; LT = 20 years denotes the lifetime of facilities; $r = 10\%$ is the discount rate accounting for the time value of money for future investment (Ertürk, 2012; Manioğlu & Yılmaz, 2006). The economic analysis is with respect to each experimental case as listed in Table 1, so that we could incorporate geopolymer production conditions into the analysis.

RESULTS AND DISCUSSION

3.1 Ash Characterization

Table 3 shows that coal fly ash contains high content of Fe_2O_3 and CaO, which may have contributed to its brown colour appearance. This is different from the general grey colour appearance as previously reported (*e.g.*, Dananjayan et al. (2016), Memon and Khan (2017) and Ukwattage et al. (2015)), which generally corresponded to higher contents of silica (SiO_2) and alumina (Al_2O_3) than Fe_2O_3 and CaO. SiO_2 and Al_2O_3 are the third and fourth richest oxide components in the coal fly ash. The original OPC contains a higher content of CaO (77.47 wt.%) and lower contents of SiO_2 and Fe_2O_3 (Please see Table S1 in the Supplemental Material).

The morphology of coal fly ash (Figure 1 (a)) shows that fly ash particles are spherical in shape,

which is consistent with existing studies (Medina et al., 2010; Siddique, 2010). The spheres could have been formed by the melting and agglomeration of the minerals in the ash (Georgakopoulos et al., 1994). The particles with irregular shapes denote the unburned material such as carbon, mineral aggregates (quartz), agglomerated particles and amorphous particles (Kutchko and Kim, 2006). The XRD pattern (Figure 1 (b)) of the ash displays peaks due to quartz, mullite, magnetite, and hematite (Álvarez-Ayuso et al., 2008). Magnetite and hematite are the mineral forms of iron oxides, while quartz and mullite prove the presence of silica and alumina.

The infrared spectrum (Figure 1 (c)) shows the main absorptions occurred at 460, 594, 795, 1104 and 1455 cm^{-1}. The two spectrum bands at 594 and 795 cm^{-1} are attributed to Si-O-Si symmetric stretching vibrations (quartz phase) (Ciocinta et al., 2012; Mucsi et al., 2016). The two main spectral bands at 460 and 1104 cm^{-1} are attributed to Si-O-Si and O-Si-O bending, and Si-O-Si and Al-O-Si asymmetric stretching, respectively (Álvarez-Ayuso et al., 2008). This is consistent with the presence of quartz and mullite as detected in the XRD analysis. The presence of absorption band at 1455 cm^{-1} should be related to the carbonate group (Santa et al., 2013).

3.2 Parametric Analysis

3.2.1 Heat resistance

Figure 2 (a) shows that the heat resistance of geopolymer is improved upon the increase in the amount of Na_2SiO_3 solution. The dry weight loss decreases by around 40% when the amount of Na_2SiO_3 solution increases from 50% to 200%. As the amount of Na_2SiO_3 solution increases, the dissolution step and the formation of the mobile precursors will be facilitated, resulting in a more complete geopolymer structure and improved heat resistance.

Figure 2 (b) shows that the dry weight loss of geopolymer firstly decreases with the increasing mole

ratio of SiO_2/Na_2O and then increased. The increase in the mole ratio of SiO_2/Na_2O is subject to the increase in the amount of Na_2SiO_3 solution and decrease in the amount of NaOH. Like the case of Na_2SiO_3 solution, the synthesis of geopolymer is firstly enhanced by the increasing Na_2SiO_3 amount for the dissolution step and the formation of precursors (Van Jaarsveld et al., 2002a). However, as the mole ratio continues to increase and the amount of NaOH continues to decrease, the pH is not high enough to ensure the dissolution of aluminosilicate particles. Furthermore, as the pH decreases, the subsequent action of hydroxide ions with the dissolved aluminate and silicate will also be adversely affected (Xu and Van Deventer, 2000).

Figure 2 (c) shows that the dry weight loss of geopolymer decreases by around 50% as the mass ratio of ash/alkaline solution increases from 2 to 3, and then remains almost constant afterwards. This suggests that the coal fly ash amount is sufficient when the ratio reaches 3 and the further increase in the ratio does not cause an apparent effect. Figures 2 (d)-(f) shows that the average heat resistance of geopolymer for the lower temperature (60 ^oC) is generally better than that for the higher temperature (80 ^oC) under the same curing time within 24 h. The average dry weight loss is around 3% for the cases of 80 and 35 ^oC and 2.5% for the case of 60 ^oC. Under the cases of lower temperature (35 and 60 ^oC), there is no significant effect of curing time on the heat resistance. Under the case of higher temperature (80 ^oC), the heat resistance improves when the curing time increases from 20 to 24 h.

3.2.2 Acid resistance

As shown in Table 4, there is no distinct trend about the variation of the weight loss of geopolymer regarding the amount of Na_2SiO_3. The variation of the weight loss as the SiO_2/Na_2O mole ratio is like the case of heat resistance: the acid resistance is firstly enhanced as the increase of the SiO_2/Na_2O mole ratio from 1 to 1.15 and weakened afterwards (1.15 to 1.45). This may have resulted from a similar mechanism as related to the variation of the amount of Na_2SiO_3 solution and pH value. The

weight loss increases as the ash/alkaline solution mass ratio increases from 2 to 4 and slightly decreases afterwards (4 to 5). The initial increase in the weight loss should be related to the higher content of iron oxide which is more soluble in acidic environments (Kumar, 2015), while the slight decrease should be an indication of the saturated effect of iron oxide. With a curing time less than 24 h, the temperature has different effects on the weight loss for the cases of 60 and 80 ^{o}C. Higher temperature and longer curing time are favorable conditions for the acid resistance. With a curing time between 2.5 and 9 days, the average weight loss for the case of 35 ^{o}C is like that for the case of 60 ^{o}C.

3.3 Comparison with OPC

Table 5 shows the heat resistance of geopolymer is better than that of OPC, and thus the high iron oxide and calcium oxide contents do not serve to deteriorate the heat resistance. OPC binder requires retaining more water of hydration within gel phases to maintain the structural integrity of the binder, which serves to lower its heat resistance. The geopolymer has a lower acid resistance than OPC. Considering that OPC has an ultra-high calcium oxide content of 77.47 wt%, this suggests that the lower acid resistance for geopolymer should be mainly caused by the high iron oxide content in the coal fly ash that could be leached out by acid immersion. The high iron oxide content in geopolymer is also confirmed by its XRD pattern that displays peaks due to magnetite and hematite as shown in Figure S2 (a) (Please see the Supplemental Material). It is worth noting that although we have the data of compressive strength in this work, the high calcium oxide content in the fly ash may lead to the formation of calcium silicate hydrate, which may serve to enhance the strength in composite geopolymers (Huang et al., 2018; Kumar et al., 2010).

3.4 Economic Analysis

3.4.1 Average NPV and components

Figure 3 (a) shows that the geopolymer production for most of the cases is hardly profitable, except for case 1, 11 and 12. These three cases correspond to a relatively high ash/alkaline solution ratio of 5, 4 and 5 (Table 1), respectively, which means a low usage of the Na_2SiO_3 solution and NaOH. Specifically, an ash/alkaline solution ratio of 4 and 5 in case 11 and 12 imply around 12 and 10 tons of daily Na_2SiO_3 solution usage as compared to around 29 and 24 tons of daily Na_2SiO_3 solution usage in case 4 and 9 with the ratio of 1.7 and 2. We further explore the detailed impact of the ash/alkaline solution ratio on the economics of geopolymer production by varying the ratio from 4 to 6 as shown in the inset figure in Figure 3 (a). It shows that the average NPV increases from around 2.5 million to 11 million USD when the ratio increases from 4 to 6.

However, additional measures need to be taken to mitigate the acid resistance issue for the geopolymer produced under the conditions of the case 11 and 12 (weight loss around 40% for both cases). The geopolymer produced under the condition of case 1 has a relatively low acid resistance (weight loss around 36%). Hence, the technical feasibility may affect the practical implementation under the conditions of case 1, 11 and 12. Geopolymer production under the condition of case 9 is not economically feasible in addition to its poor technical feasibility regarding heat resistance.

Figure 3 (b) shows that the geopolymer selling dominates the benefit components and is about triple of the income from the refuse disposal fee collection. This is because the price of geopolymer is around 100% higher than the refuse disposal fee and the mass of geopolymer is 9-20% larger than that of the coal fly ash collected. There are no big differences in the benefit components among the different cases. The Na_2SiO_3 cost dominates the cost components followed by the NaOH cost, except for case 1 where the costs of Na_2SiO_3 and NaOH are comparable. The significant smaller Na_2SiO_3 costs in Figure 3 (b) correspond to the high NPVs in Figure 3 (a) for case 1, 11 and 12, while the much higher Na_2SiO_3 cost (around 2.5 times of case 12) for case 4 in Figure 3 (b) corresponds to the

large negative NPV in Figure 3 (a). Other components such as the equipment cost, electricity and water cost, and the capital cost play a minor role in affecting the overall economics. Note that the costs of land, construction, O&M, electricity, water, transport, geopolymer, and refuse disposal are highly contingent upon the countries or regions applied.

3.4.2 NPV distributions

Figure 4 shows that the NPV distributions shift to the left from case 1 to case 4 corresponding to the increased use of the Na_2SiO_3 solution. The profitability chance decreases from around 93% for case 1 to 0% for case 4 that has an all-negative NPV distribution. For batch 2 cases, the NPV distribution shifts slightly to the right from case 5 to case 8. This corresponds to a slight decrease in the productivity of geopolymer from around 64 to 63 tons per day, leading to the decrease in the geopolymer income. The profitability chance ranges from 7% for case 5 to 12% for case 8. For batch 3 cases, the NPV distributions shift to the right as the daily use of Na_2SiO_3 solution decreases from around 24 tons for case 9 to 9.7 tons for case 12. The profitability chance increases from 0% for case 9 to 98% for case 12. For batch 4-6, the benefit and cost components of the cases are similar to each other except that case 13-16 have more sets of ovens as shown in Figure 3 (b). The cost of ovens does not play a major effect on the NPV and thus the NPV distributions almost overlap with each other for case 13-24 with a consistent profitability chance around 10%.

3.4.3 NPV contours

Considering the dominant roles of geopolymer selling and Na_2SiO_3 use in the overall economic feasibility, it is desirable to explore the potential impacts of varying the prices of geopolymer and Na_2SiO_3 on the NPV of geopolymer production. Figure 5 shows the NPV contours with respect to the prices of geopolymer (50 to 250 USD/ton) and Na_2SiO_3 (300 to 900 USD/ton) under the condition of

case 12. 5% NPV denotes the NPV value that is larger than 5% NPV values in the NPV distribution, while 95% NPVs denote the NPV value that is larger than 95% NPV values in the NPV distribution.

In Figure 5 (a) the zero NPV contour is an indication of the economic boundary. Above this contour, there is more than 50% of the chance for the geopolymer production to be profitable, while below contour, there is less than 50% of the change to be profitable. When the price of geopolymer is higher than around 120 USD/ton, the profitability chance of the production is always higher than 50% for the Na_2SiO_3 price up to 900 USD/ton. However, when the price of geopolymer decreases to around 50 USD/ton, the Na_2SiO_3 price must be lower than around 480 USD/ton to make the profitability chance higher than 50%. Under the geopolymer price of 250 USD/ton, the average NPV could be around 45 and 25 million under the Na_2SiO_3 prices of 300 and 800 USD/ton.

In Figure 5 (b), above the zero NPV contour, the geopolymer production is profitable, from a statistical point of view. Figure 5 (b) shows that the geopolymer production will be profitable even if the geopolymer price is as low as 50 USD/ton when the Na_2SiO_3 price is around 400 USD/ton. When the Na_2SiO_3 price increases to 800 USD/ton, the production is profitable only if the geopolymer price is higher than 130 USD/ton. When the geopolymer price is as high as 250 USD/ton, the production is surely profitable with the NPV larger than 20 million under the Na_2SiO_3 price of 800 USD/ton. Similarly, in Figure 5 (c) regarding the 95% NPV, the geopolymer production is not profitable if it is below the zero NPV line. When the geopolymer price is lower than 50 USD/ton, the production is not profitable when the Na_2SiO_3 price is higher than around 560 USD/ton.

CONCLUSIONS

The heat resistance of geopolymer was better than OPC. However, the high iron oxide content made the acid resistance of geopolymer inferior to OPC. The economics of geopolymer production changes

significantly upon the variation in the arrangement of material use and geopolymer price. The costs of Na_2SiO_3 and NaOH and the benefit of geopolymer selling were major factors affecting the economic feasibility of geopolymer production. The results of economic analysis serve as the basis for policymakers and investors to make informed decisions about practical implementation of geopolymer production. The method of this work can be used to accumulate a data bank on the economic feasibility conditions of different types of geopolymers to facilitate the decision-making process of policy-makers and investors.

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